

High-temperature study of superconducting hydrogen and deuterium sulfide

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Hydrogen-rich compounds are extensively explored as candidates for a high-temperature superconductors. Currently, the measured critical temperature of 203 K in hydrogen sulfide (H_3S) is among the highest over all-known superconductors. In present paper, using the strong-coupling Eliashberg theory of superconductivity, we compared in detail the thermodynamic properties of two samples containing different hydrogen isotopes H_3S and D_3S at 150 GPa. Our research indicates that it is possible to reproduce the measured values of critical temperature 203 K and 147 K for H_3S and D_3S by using a Coulomb pseudopotential of 0.123 and 0.131, respectively. However, we also discuss a scenario in which the isotope effect is independent of pressure and the Coulomb pseudopotential for D_3S is smaller than for H_3S . For both scenarios, the energy gap, specific heat, thermodynamic critical field and related dimensionless ratios are calculated and compared with other conventional superconductors. We shown that the existence of the strong-coupling and retardation effects in the systems analysed result in significant differences between values obtained within the framework of the Eliashberg formalism and the prediction of the Bardeen-Cooper-Schrieffer theory.

Keywords: Superconductors; Hydrogen-rich compounds; High-pressure; Thermodynamic properties

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I. INTRODUCTION

Hydrogen sulfide at high pressure (p) is believed to be a phonon-mediated superconductor characterized by the highest superconducting transition temperature (T_C) ever measured. The recent electrical, resistance, magnetic susceptibility and Raman spectra measurements performed using a diamond anvil cell by Drozdov *et al.* showed that H_2S transforms into a metal and then into a superconductor around 90–100 GPa [1, 2]. In a sample prepared at $p < 150$ GPa and at low temperature of 100–150 K, T_C increases with pressure from 31 K at 115 GPa to 60 K at 145 GPa. Then, as pressure increases above 150 GPa a drastic increase of T_C was observed and finally T_C reaches 150 K at 200 GPa [1]. The above experiment was directly inspired by previous theoretical calculations conducted within the density functional perturbation theory by Li *et al.* [3]. It should be noted that the measured results for $p < 150$ GPa are in general agreement with the theoretical predictions mentioned. Additional experimental measurements on a sample prepared at $p > 150$ GPa and at high temperature of 220–300 K found superconductivity with T_C as high as 203 K, breaking all the records thus far [1].

These differences between samples prepared at low/high temperature and pressure are probably associated with the dissociation of solid hydrogen sulfide. Theoretical prediction clearly suggests the pressure-induced

decomposition of H_2S to H_3S and elemental sulfur according to the following energetically allowed scheme: $3\text{H}_2\text{S} \rightarrow 2\text{H}_3\text{S} + \text{S}$ [4–6]. It means that H_3S , above 50 GPa remains the more stable stoichiometry than H_2S , which suggests that the highest critical temperatures observed experimentally come from H_3S compound [6, 7].

This discovery has stimulated significant interest in studying the underlying superconducting mechanism [6, 8–12] and searching for novel methods to decrease the external hydrostatic pressure and increase the critical temperature of dense hydrogen sulfide [13]. The systematic theoretical study performed in paper [14] showed that the maximum value of the critical temperature in the family of the H_nS -type compounds ($n \geq 2$) can even be equal to ~ 290 K. According to the results of this work, hydrogen sulfide is a candidate for a high temperature superconductor with a critical temperature as high as the room temperature.

Motivated by the recent experimental and theoretical progress in this area, we have carried out calculations to explore in detail the thermodynamic properties of superconducting hydrogen sulfide H_3S and deuterium sulfide D_3S at high pressure. In particular, we chose a pressure equal to ~ 150 GPa due to the fact that the experimental results exist for both systems. Moreover, T_C for H_3S is close to the maximum measured value. The very large values of electron-phonon coupling strength $[\lambda]_{\text{H}_3\text{S}} = 2.067$ and $[\lambda]_{\text{D}_3\text{S}} = 2.065$ calculated using *ab*

initio methods [15], caused that our investigations were conducted within the framework of the strong-coupling Eliashberg theory, which allows us to describe the thermodynamic properties of conventional superconductors with experimental accuracy.

The paper is organized as follows. Section II contains a short outline of the strong-coupling Eliashberg formalism. In Section III, we discuss and compare the thermodynamic properties of superconducting H₃S and D₃S systems at 150 GPa. Section IV summarizes the obtained results.

II. ELIASHBERG FORMALISM

The finite temperature Eliashberg equations for the superconducting energy gap $\Delta(\omega) \equiv \phi(\omega)/Z(\omega)$ and the mass renormalization function $Z(\omega)$ written in a mixed representation (formulated of both the real and imaginary frequency axis) are given by [16]:

$$\begin{aligned} \phi(\omega) = & \frac{\pi}{\beta} \sum_{m=-M}^M \frac{[\lambda(\omega - i\omega_m) - \mu^* \theta(\omega_c - |\omega_m|)]}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} \phi_m \quad (1) \\ & + i\pi \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[[N(\omega') + f(\omega' - \omega)] \right. \\ & \times K(\omega, -\omega') \phi(\omega - \omega') \left. \right] \\ & + i\pi \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[[N(\omega') + f(\omega' + \omega)] \right. \\ & \times K(\omega, \omega') \phi(\omega + \omega') \left. \right] \end{aligned}$$

and

$$\begin{aligned} Z(\omega) = & 1 + \frac{i\pi}{\omega\beta} \sum_{m=-M}^M \frac{\lambda(\omega - i\omega_m) \omega_m}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} Z_m \quad (2) \\ & + \frac{i\pi}{\omega} \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[[N(\omega') + f(\omega' - \omega)] \right. \\ & \times K(\omega, -\omega') (\omega - \omega') Z(\omega - \omega') \left. \right] \\ & + \frac{i\pi}{\omega} \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[[N(\omega') + f(\omega' + \omega)] \right. \\ & \times K(\omega, \omega') (\omega + \omega') Z(\omega + \omega') \left. \right], \end{aligned}$$

where

$$K(\omega, \omega') \equiv \frac{1}{\sqrt{(\omega + \omega')^2 Z^2(\omega + \omega') - \phi^2(\omega + \omega')}}. \quad (3)$$

Here, $\beta = 1/k_B T$, symbol μ^* means the screened Coulomb repulsion, $f(\omega)$ and $N(\omega)$ are the Fermi-Dirac and Bose-Einstein distribution functions. Symbols θ and ω_c denote the Heaviside function and the cut-off frequency, respectively. For ω_c we chosen ten times the

maximum phonon frequency: $\omega_c = 10\Omega_{\max}$. The imaginary axis functions $\phi_n \equiv \phi(i\omega_n)$ and $Z_n \equiv Z(i\omega_n)$ are given by [17]:

$$\phi_n = \frac{\pi}{\beta} \sum_{m=-M}^M \frac{\lambda(i\omega_n - i\omega_m) - \mu^*(\omega_m)}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} \phi_m \quad (4)$$

and

$$Z_n = 1 + \frac{1}{\omega_n} \frac{\pi}{\beta} \sum_{m=-M}^M \frac{\lambda(i\omega_n - i\omega_m)}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} \omega_m Z_m. \quad (5)$$

The Matsubara frequency is defined as $\omega_n \equiv (\pi/\beta)(2n-1)$ where $n = 0, \pm 1, \pm 2, \dots, \pm M$, and $M = 1100$. The pairing kernel for the electron-phonon interaction is given by:

$$\lambda(z) \equiv 2 \int_0^{\Omega_{\max}} d\omega \frac{\omega}{\omega^2 - z^2} \alpha^2 F(\omega), \quad (6)$$

The Eliashberg spectral function $\alpha^2 F(\omega)$ is expressed as a sum over the contributions from scattering processes which connect electrons through phonons on the Fermi surface. Our investigations are based on the spectral function determined by using the *ab initio* plane-wave pseudopotential calculations in paper [15]:

$$\begin{aligned} \alpha^2 F(\omega) = & \frac{1}{N(0)} \sum_{\nu \mathbf{q} n n' \mathbf{k}} |g_{\nu \mathbf{q}}^{n \mathbf{k} + \mathbf{q}, n' \mathbf{k}}|^2 \delta(\xi_{n \mathbf{k} + \mathbf{q}}) \quad (7) \\ & \times \delta(\xi_{n' \mathbf{k}}) \delta(\omega - \omega_{\nu \mathbf{q}}), \end{aligned}$$

where $N(0)$, $g_{\nu \mathbf{q}}^{n \mathbf{k} + \mathbf{q}, n' \mathbf{k}}$ and $\omega_{\nu \mathbf{q}}$ denote the density of states at the Fermi energy, the electron-phonon matrix element and the phonon frequency, respectively.

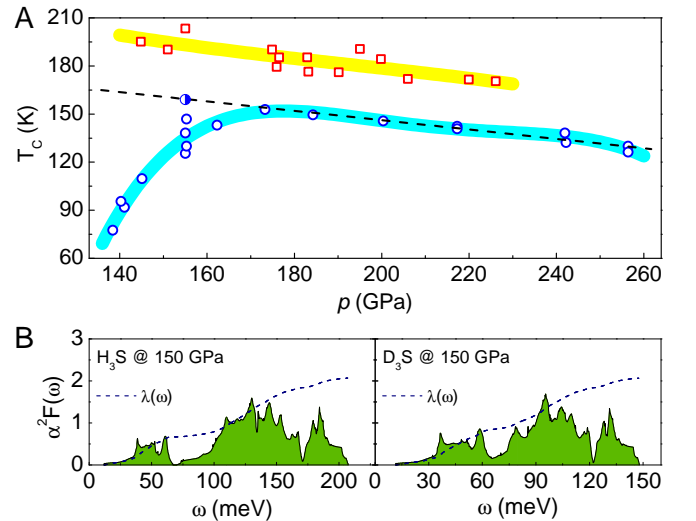


FIG. 1: (A) The pressure dependence of critical temperature for H₃S (open squares) and D₃S (open circles) [1]. The dashed line and half-filled circle represent our prediction related to the constant value of isotope effect. (B) The Eliashberg spectral functions $\alpha^2 F(\omega)$ and the electron-phonon integrals $\lambda(\omega)$ of H₃S and D₃S at 150 GPa [15].

The shape of the Eliashberg functions for H₃S and D₃S is presented in Fig. 1 (B). Moreover, the dashed line is the integration of the electron-phonon coupling strength as a function of the phonon frequency:

$$\lambda(\omega) = 2 \int_0^{\Omega_{\max}} \frac{\alpha^2 F(\omega)}{\omega} d\omega. \quad (8)$$

The main part of Fig. 1 shows the experimental values of critical temperature for H₃S (open squares) and D₃S (open circles) as a function of pressure [1]. Both from the experimental and theoretical point of view, especially interesting seems to be non-conventional behaviour of the isotope effect associated with low values of critical temperature for D₃S at low pressures. This can result from inaccuracy of measurement, instability of the system, or it can be a new effect, not observed until now. Therefore, we discuss two scenarios. The first assuming the correctness of the experimental results, $T_C = 203$ K for H₃S and $T_C = 147$ K for D₃S [1], and the second assuming invariance of the isotope effect, $T_C = 203$ K for H₃S and $T_C = 159$ K for D₃S (half-filled circle in Fig. 1 (A)).

In order to determine the all significant thermodynamic properties of H₃S and D₃S systems, we use a standard Matsubara technique to solve the Eliashberg equations. In particular, we conducted numerical calculations based on a self-consistent iteration methods [18], which were used successfully in our previous papers [19–21].

III. THE RESULTS AND DISCUSSION

The isotope effect of superconducting critical temperature is best described in terms of the isotope effect coefficient (α) defined by the relation:

$$\alpha = -\frac{\ln[T_C]_{D_3S} - \ln[T_C]_{H_3S}}{\ln[M]_D - \ln[M]_H}, \quad (9)$$

where $[M]_H$ and $[M]_D$ are the atomic mass of hydrogen and deuterium, respectively. In the case of experimental results for hydrogen and deuterium sulfide at $p = 150$ GPa we have $\alpha = 0.47$. This value is very close to the BCS value ($[\alpha]_{BCS} \approx 0.5$). However, if we take into account $T_C = 159$ K for D₃S, then $\alpha = 0.35$, which is in a general agreement with the results obtained for a higher pressures.

To study the thermodynamic properties on the quantitative level in the framework of the Eliashberg formalism we must first determine the critical value of the Coulomb pseudopotential μ_C^* . For this purpose, in the Eliashberg equations we assumed that $T = T_C$ and then we increased the value of the parameter μ^* until we reached the equality $\Delta_{m=1} = 0$ at $\mu^* = \mu_C^*$. The full dependence of $\Delta_{m=1}(\mu^*)$ are presented in Fig. 2. On this basis we can notice that for the investigated systems, Coulomb pseudopotentials, correlated with experimental

results take a typical critical values, $\mu_C^* = 0.123$ for H₃S and $\mu_C^* = 0.131$ for D₃S. However, untypical is the fact that $[\mu_C^*]_{H_3S} < [\mu_C^*]_{D_3S}$, because according to the Morel-Anderson formula [22]:

$$\mu^* = \frac{\mu}{1 + \mu \ln \left(\frac{\omega_{eL}}{\omega_{ln}} \right)} \simeq \frac{1}{\ln \left(\frac{\omega_{eL}}{\omega_{ln}} \right)}, \quad (10)$$

μ^* is proportional to the average phonon frequency (ω_{ln}) which relates to the dynamics of the superconducting state: $\omega_{ln} \equiv \exp \left[\frac{2}{\lambda} \int_0^{+\infty} d\Omega \frac{\alpha^2 F(\Omega)}{\Omega} \ln(\Omega) \right]$. Due to the atom mass the average phonon frequency is larger for the system containing hydrogen than for the one containing deuterium, so we should observe that $[\mu_C^*]_{H_3S} > [\mu_C^*]_{D_3S}$. This situation is possible when we take into account $T_C = 159$ K for D₃S. Then, $[\mu_C^*]_{D_3S} = 0.088$. Such a small value of μ_C^* is physically possible in the hydrogen rich materials, as was confirmed by the recent research of platinum hydride [23].

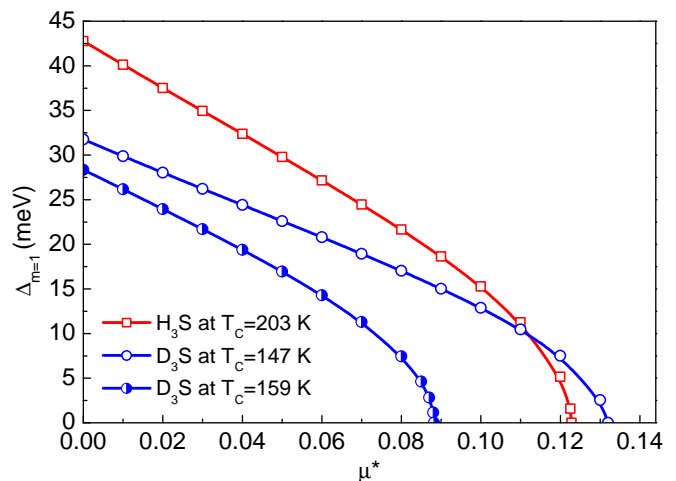


FIG. 2: The full dependence of the maximum value of the order parameter on the Coulomb pseudopotential.

Let us notice, that for the above values of μ_C^* , the critical temperatures observed in the experiment cannot be correctly reproduced using the analytical formulas for T_C . In particular, the Allen-Dynes expression [24] predicts that $[T_C(\mu_C^*)]_{H_3S} = 176$ K, $[T_C(\mu_C^*)]_{D_3S}^{(1)} = 130$ K and $[T_C(\mu_C^*)]_{D_3S}^{(2)} = 147$ K; while the McMillan formula [25] gives even more inaccurate results: $[T_C(\mu_C^*)]_{H_3S} = 148$ K, $[T_C(\mu_C^*)]_{D_3S}^{(1)} = 110$ K and $[T_C(\mu_C^*)]_{D_3S}^{(2)} = 121$ K. It should be noted that in order to improve readability of this paper we use in the superscript the abbreviation (1) in the context of the first scenario ($T_C = 147$ K for D₃S) and abbreviation (2) in the context of the second scenario ($T_C = 159$ K for D₃S) discussed in this paper.

By using the Eliashberg spectral functions and the determined above Coulomb pseudopotentials as an input parameters to the Eliashberg equations, we obtained the

superconducting energy gap values $\Delta(0) = 42.74$ meV for H_3S and $\Delta(0)^{(1)} = 31.14$ meV and $\Delta(0)^{(2)} = 34.21$ meV for D_3S , which corresponds to $[2\Delta(0)/k_B T_C]_{\text{H}_3\text{S}} = 4.89$, $[2\Delta(0)/k_B T_C]_{\text{D}_3\text{S}}^{(1)} = 4.92$ and $[2\Delta(0)/k_B T_C]_{\text{D}_3\text{S}}^{(2)} = 4.99$.

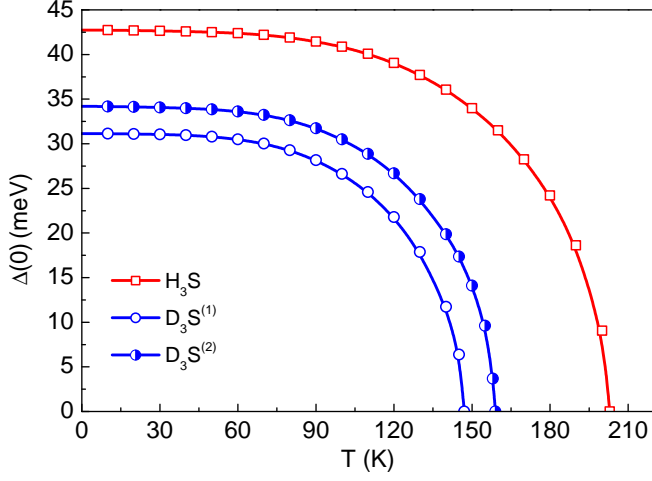


FIG. 3: The full dependence of the superconducting energy gap on the temperature.

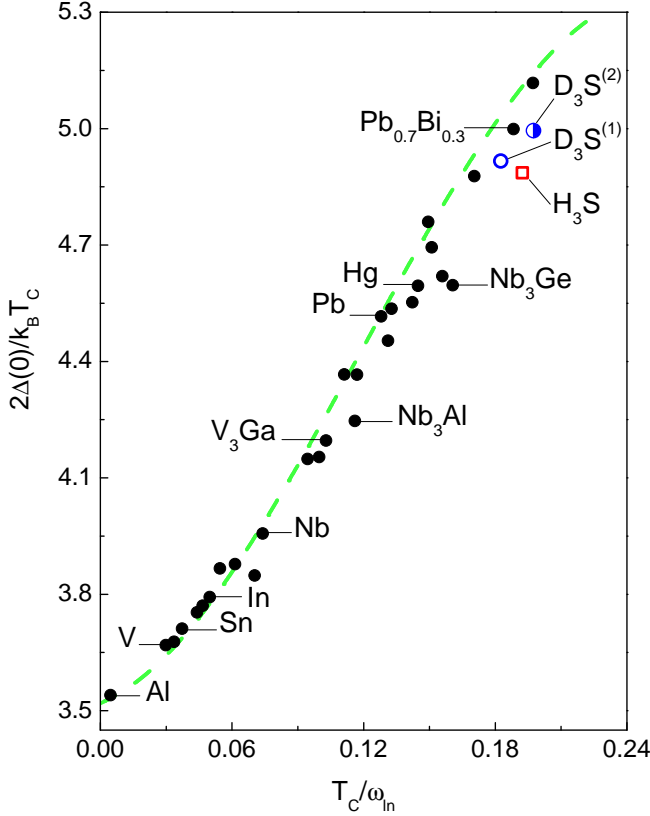


FIG. 4: The superconducting gap ratio as a function of T_C/ω_{ln} . The results for the selected conventional superconductors are taken from paper [26]. The dashed line are obtained on the basis of paper [27].

The temperature dependences of the superconducting gaps are shown in Fig. 3. Moreover, we compared the calculated gap ratio for H_3S and D_3S with other conventional superconductors. In particular, in Fig. 4 we can see $2\Delta(0)/k_B T_C$ as a function of the strong-coupling index T_C/ω_{ln} [28]. At this point it should be noted that in the weak coupling BCS limit $T_C/\omega_{ln} \rightarrow 0$. In Fig. 4, the full circles representing results for selected conventional superconductors and dashed line are taken from papers [26, 27]. It is clearly visible that although the obtained results differ significantly from the prediction of the classic BCS theory ($[2\Delta(0)/k_B T_C]_{\text{BCS}} = 3.53$), they are close to the trend determined by the conventional superconductors. The recent study reported by Nicol and Carbotte [29] generally confirmed our calculations.

In the next step, we calculated the specific heat difference between the superconducting and the normal states:

$$\frac{\Delta C}{k_B N(0)} = -\frac{1}{\beta} \frac{d^2 [\Delta F/N(0)]}{d(k_B T)^2}, \quad (11)$$

where symbol ΔF denotes the free energy difference between the superconducting and the normal state [30]:

$$\begin{aligned} \frac{\Delta F}{N(0)} = & -\frac{2\pi}{\beta} \sum_{n=1}^M \left(\sqrt{\omega_n^2 + \Delta_n^2} - |\omega_n| \right) \\ & \times \left(Z_n^S - Z_n^N \frac{|\omega_n|}{\sqrt{\omega_n^2 + \Delta_n^2}} \right). \end{aligned} \quad (12)$$

Here, Z_n^S and Z_n^N are the mass renormalization functions for the superconducting and for the normal state, respectively. Let us note that the Eq.(12) can be easily computed, once the imaginary axis Eliashberg equations (4) and (5) are solved.

The specific heat difference for H_3S and D_3S as a function of temperature was shown in Fig. 5. The characteristic specific heat jump at T_C was marked by vertical

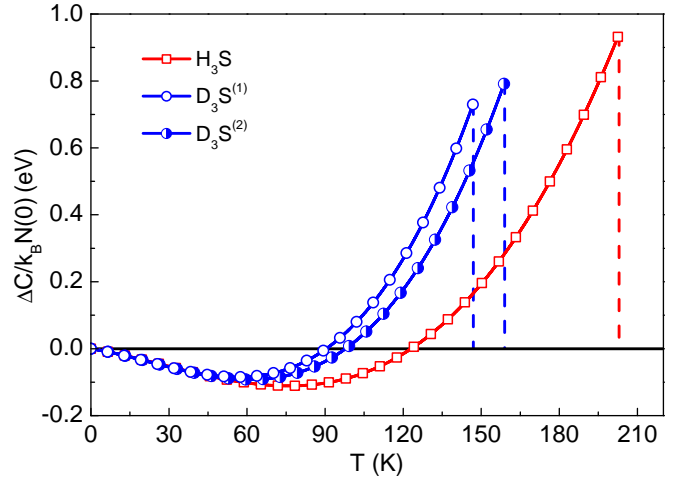


FIG. 5: The specific heat difference as a function of temperature for H_3S and D_3S at 150 GPa.

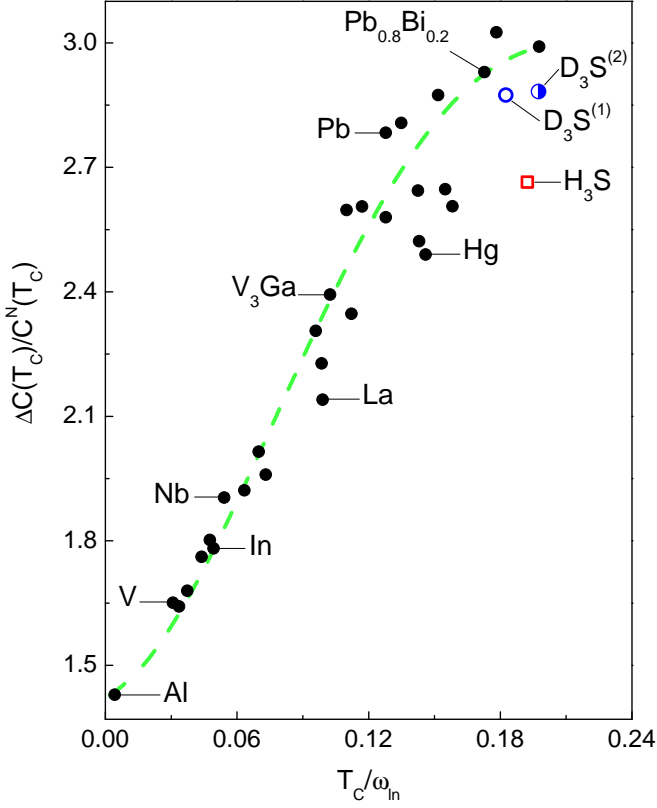


FIG. 6: The specific heat ratio as a function of T_C/ω_{ln} . The results for the selected conventional superconductors are taken from paper [26]. The dashed line are obtained on the basis of paper [31]. For H_3S , $D_3S^{(1)}$ and $D_3S^{(2)}$ the ratio $\Delta C(T_C)/C^N(T_C)$ is equal to 2.66, 2.87 and 2.88, respectively.

dashed lines. At critical temperature, the BCS result for the dimensionless ratio $\Delta C(T_C)/C^N(T_C)$ is equal to 1.43, and is universal, like the superconducting gap ratio. In the above ratio, the specific heat for the normal state is defined as: $C^N = \gamma T$, where symbol γ denotes the Sommerfeld constant: $\gamma \equiv (2/3)\pi^2(1 + \lambda)k_B^2 N(0)$. The comparison of the specific heat ratio as a function of T_C/ω_{ln} for the investigated systems with other conventional superconductors looks similar to the gap ratio (see Fig. 6). However, despite the fact that the value calculated for D_3S at 150 GPa is very close to the dashed line adopted from paper [31], the value for H_3S clearly departs from this trend.

In the next step, on the basis of Eq.(12), the thermodynamic critical field H_C and the deviation function of the thermodynamic critical field D can be obtained from the following relations:

$$\frac{H_C}{\sqrt{N(0)}} = \sqrt{-8\pi[\Delta F/N(0)]} \quad (13)$$

and

$$D = \frac{H_c(T)}{H_c(0)} - \left[1 - \left(\frac{T}{T_c}\right)^2\right]. \quad (14)$$

The influence of the temperature on the thermodynamic critical field was presented in Fig. 7 (A). In Fig. 7 (B) we supplement our results with the calculated thermodynamic critical field deviation as a function of the reduced temperature. The results predicted by the weak-coupling BCS theory are presented with a dashed line [32]. As for the investigated materials our results confirm the non-BCS behaviour of the thermodynamic critical field deviation. In the BCS theory, the dimensionless ratio connected with thermodynamic critical field $T_C C^N(T_C)/H_C^2(0)$ also takes the universal value equal to 0.168. In the case of hydrogen and deuterium sulfide we observed a high derogation from the BCS theory prediction, in particular $[T_C C^N(T_C)/H_C^2(0)]_{H_3S} = 0.130$, $[T_C C^N(T_C)/H_C^2(0)]_{D_3S}^{(1)} = 0.127$ and $[T_C C^N(T_C)/H_C^2(0)]_{D_3S}^{(2)} = 0.126$. The comparison with other superconductors is presented in Fig. 8. Also here the result computed for H_3S clearly differs from the general trend set by the dashed line [31].

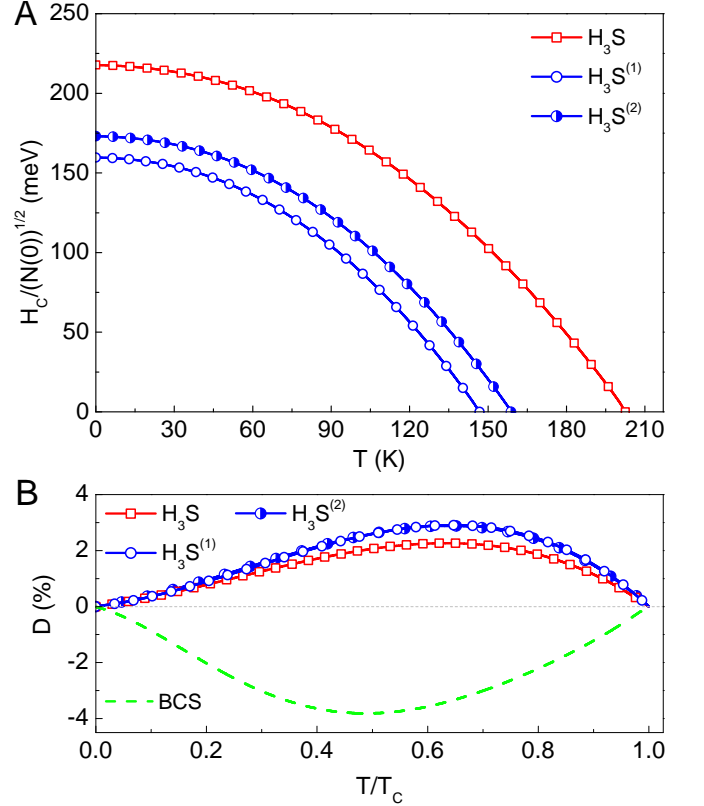


FIG. 7: (A) The thermodynamic critical field and (B) the critical field deviation as a function of temperature for H_3S and D_3S at 150 GPa.

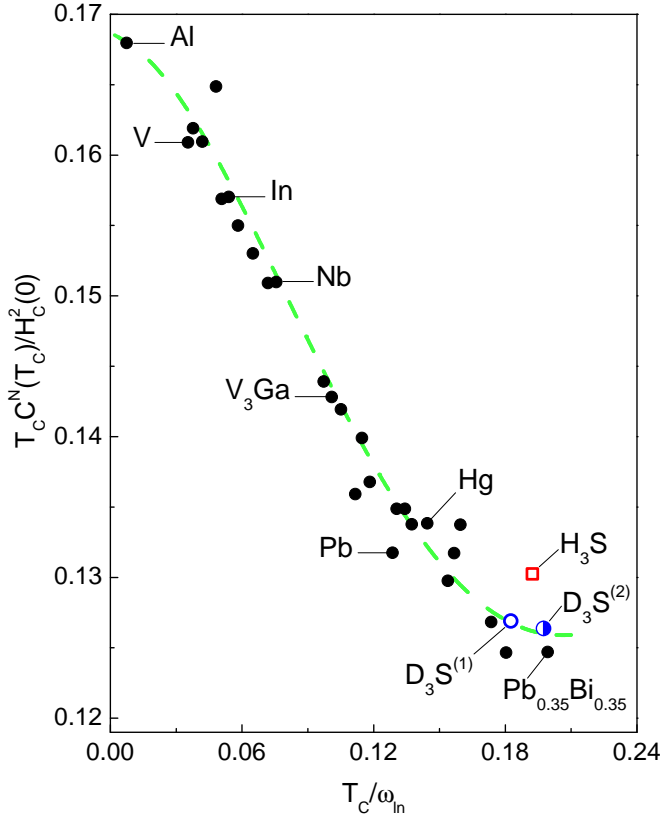


FIG. 8: The thermodynamic critical field ratio as a function of T_C/ω_{in} . The results for the selected conventional superconductors are taken from paper [26]. The dashed line are obtained on the basis of paper [31].

In the last step, we computed the London penetration depth (λ_L):

$$\frac{1}{e^2 v_F^2 N(0) \lambda_L^2(T)} = \frac{4\pi}{3\beta} \sum_{n=1}^M \frac{\Delta_n^2}{Z_n^S [\omega_n^2 + \Delta_n^2]^{3/2}}, \quad (15)$$

where e is the electron charge and v_F is the Fermi ve-

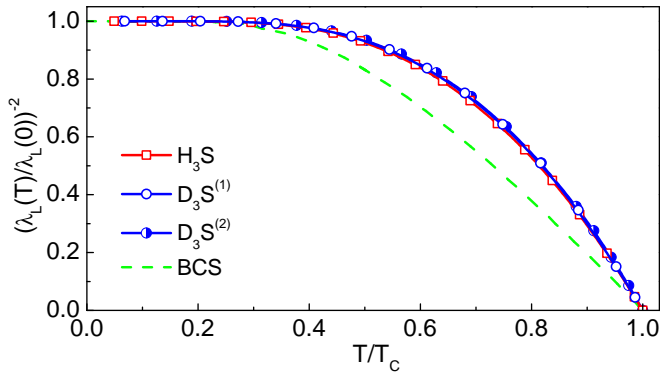


FIG. 9: The normalized London penetration depth as a function of temperature.

locity [26]. The normalized λ_L^{-2} as a function of temperature is presented and compared with the BCS theory prediction in Fig. 9.

The differences observed between the BCS and the Eliashberg theory are connected with the fact that the BCS approach does not include the strong coupling corrections and the retardation effect of the actual electron-phonon interactions as described by the Eliashberg theory.

IV. CONCLUSIONS

In this paper, using the phonon-mediated Eliashberg theory, we have investigated the thermodynamic properties of superconducting hydrogen and deuterium sulfide at high pressure ($p = 150$ GPa). In particular, the energy gap, the specific heat, the thermodynamic critical field and the London penetration depth were calculated for Coulomb pseudopotentials $[\mu_C^*]_{H_3S} = 0.123$ and $[\mu_C^*]_{D_3S}^{(1)} = 0.131$ determined on the basis of experimental values of critical temperature ($[T_C]_{H_3S} = 203$ K, $[T_C]_{D_3S}^{(1)} = 147$ K) and $[\mu_C^*]_{D_3S}^{(2)} = 0.088$ determined on the basis of our prediction of critical temperature for deuterium sulfide ($[T_C]_{D_3S}^{(2)} = 159$ K). Next, on the basis of the calculated thermodynamic functions, it was proven that the values of the dimensionless ratios $2\Delta(0)/k_B T_C$, $\Delta C(T_C)/C^N(T_C)$ and $T_C C^N(T_C)/H_C^2(0)$ differ significantly from the predictions of the BCS theory. These discrepancies arise from the existence of the strong-coupling and retardation effects in the systems investigated. The Eliashberg theory goes beyond the BCS theory to include these effects.

The results presented in this paper, are expected to stimulate experimental and theoretical exploration and discovery of new superconducting hydrogen-containing materials like H_3S . Moreover we hope that future measurements will be able to determine conclusively the pressure dependence of critical temperature for hydrogen/deuterium sulfide.

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